Final Technical Report
for

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**Generation and Characterization of Attosecond Pulses**
**DOE Grant Number: DE-FG02-01ER15156**
**2002 – 2005 Final report for Rochester Sub-Project**

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*Executive Summary*

The research undertaken in this project has been directed toward the area of attoscience, in particular the problem of attosecond metrology. That is, the accurate determination of the electric field of attosecond XUV radiation. This outstanding problem has been identified as a critical technology for further development of the field, and our research adds to the area by providing the first method for characterization using the harmonic radiation itself as a tool.

The technical effectiveness of this approach is very high, since it is vastly easier to detect XUV radiation directly than the via the spectrum of photoelectrons liberated from atoms by it. This means that the experimental data rate can be much higher in principle using all-optical detection that electron detection, which will greatly aid the utility of harmonic XUV sources in attoscience applications.

There are as yet no direct public benefits from this area of scientific research, though access to material structural dynamics on unprecedented brief timescales are expected to yield significant benefits for the future.

*Goals and Accomplishments*

During the course of the project we made significant progress toward the primary goals of the project. In particular, several new methods for XUV metrology were proposed, and work continues to implement these experimentally. In addition, some new ideas for enhanced nonlinear responses for XUV fields were explored both theoretically and experimentally. A complementary effort on source development was undertaken by Dr. L. DiMauro at Brookhaven National Laboratory (now at Ohio State University) and together these components formed a strong collaborative effort.

*Summary of Project Activities*

Specifically, we have developed concepts for several new methods for the complete characterization of attosecond XUV pulses, and have obtained experimental results related to several novel nonlinearities that we believe will scale into this wavelength and pulse duration regime. A number of papers have been submitted on this

work, and several reports have been given at conferences. Indeed, one of us (IAW) has given invited talks on "Attosecond Metrology" at CLEO in 2003 and at the OSA/ILS-XVII meeting in 2005.

Two University of Rochester students worked on this project: Ellen Kosik and Vincent Wong. Mr. Wong completed his Ph.D. degree in 2004, supervised by one of the co-PIs (RWB). Ms. Kosik completed her Ph.D. degree in 2004, after undertaking experimental work at the University of Oxford, UK, with one of the co-PIs (IAW). Both students have contributed to several publications and conference proceedings.

The BNL collaboration was very productive, resulting in two joint papers. There have been a number of personnel visits between the groups. Ellen Kosik, spent one month during the summer of 2002 at BNL. She assisted SUNY-Stony Brook graduate student Philip Colosimo, with the implementation of the new pulse diagnostic for the BNL laser system, and developed an understanding of the experimental issues associated with photoelectron spectrometry. It is intended that a similar exchange will take place this year. Dr. DiMauro has visited Oxford to discuss the project, and to present a seminar on the BNL laser source.

We have also continued collaborations with Dr. E. Cormier at CELIA, Bordeaux, France and Dr. C. Dorrer of Bell Labs/Lucent Technologies, Holmdel, NJ.

A major development, which had significant implications for this project, is the initiation of collaboration with ICSTM, Rutherford Appleton, reading, Birmingham and Newcastle Universities under the EPSRC Basic Technology program. Dr. DiMauro is a named collaborator on the project. This grant continues to fund the project at Oxford over the until 2007, providing equipment, personnel and operating and travel funds to sustain the collaboration with BNL and with Rochester. In addition, the Oxford group and BNL (now Ohio State) group are members of the European Research Training Network XTRA, which will also support some collaborative activity. We expect to continue to foster and develop these links via personnel visits, student exchanges and joint publications, beyond the completion of this present grant.

The details of the two major thrusts of the Rochester portion of this project are summarized in the next two sections. The report concludes with a summary of the publications and conference reports relating to work supported by this grant.

Novel techniques for attosecond pulse measurement

The main effort in the past year has been the exploration of several new methods for attosecond XUV pulse characterization using both photoelectron spectral interferometry as well as interferometry directly in the XUV. The main goals were to explore novel methods for determining the pulse shape of the electric field of XUV attosecond bursts generated by high-harmonic emission from atomic gases, that would enable simple, robust and rapid phase recovery. In two papers we analysed the possibilities of implementing both spectral shearing interferometry and simplified chronocyclic

tomography using known techniques of accerlating electrons liberated by XUV radiation from the target atoms in an additional infrared field. These methods look feasible, though they are yet to be implemented experimentally.

A second approach was to examine the possibility to make measurements directly in the XUV, since the signal to noise is likely to be very much higher than for photoelectron spectroscopy, and the spectrometer resolution requirements are not so onerous. We devised a simple method, whose detailed analysis showed great promise.(Cormier et al 2005) In fact it turned out that a group at CEA Saclay had come up with the same idea simultaneously, and were able to implement a simple version, verifying our analysis in the process. We have since devised a scheme suited to individual attosecond pulses, and have published three papers detailing the theory and proposed experimental implementation of these two new methods of using spectral phase interferometry to characterise attosecond pulses in the XUV, via spatial and spectral encoding.(Kosik et al, 2005 a, 2005 b) As part of the experimental development we have spent 2 weeks working on the CELIA laser in Bordeaux testing preliminary ideas on using an acousto-optic shaper to produce two sheared pulses in a CPA laser.

We also developed some new methods for the characterization of pulses of one or two optical cycles in duration, that operated at the Shannon sampling limit, and were therefore useful for extremely spectrally broad pulses measured with relatively low resolution spectrometers.(Kosik et al 2004, Wyatt et al, 2006) This class of measurements are important, because this is the sort of pulse that is used to drive high-harmonic generation in the true isolated attosecond pulse regime, and the XUV pulse character is determined entirely by the optical driving pulse character. In collaboration with the MBI group in Berlin we have tested these methods, and a new set of experiments are scheduled in April 2006. These experiments will investigate the ability to generate temporally and spectrally sheared replicas and to make XUV SPIDER measurements out of a filament broadening device. The ability to rapidly and reliably measure sub-5fs pulses is a critical enabling technology for high-harmonic generation.

Strong field enhanced nonlinearities for XUV metrology

This portion of the research was successfully completed and led both to some anticipated and some unexpected results. The goal of this work was to perform proof-of-principle studies of the ability to transfer modulation from one laser field at one frequency to another laser field at a different frequency. The ability to perform a transfer of this sort is crucial to the use of SPIDER techniques for the analysis of the time-dependent spectral content of an x-ray laser field, because convenient optical-frequency measurements could be used to analyze the x-ray field. Studies of this sort provided the basis of the PhD thesis of Vincent Wong, who was supported by this program. Our approach was to make use of electromagnetically induced transparency (EIT) to facilitate this transfer process. Our work began with a detailed study of the nature of EIT resonances in a sodium atomic vapor (Wong et al., 2003). The results of this work allowed us to choose optimal conditions for implementing the transfer of modulation, as reported by Bennink et al. (2005). In the process of investigating the process of transfer to modulation by means of EIT, we uncovered some subtle features of the nature of the nonlinear optical response that were reported in several publications (Agarwal and Boyd, 2003; Boyd et al, 2004;

Rzazewski and Boyd, 2004; Milonni and Boyd, 2004). We also developed new techniques for the characterization of the nonlinear optical response of materials to ultrashort laser pulses (Piredda et al. 2005). Furthermore, we performed some studies aimed at determining the stability of laser beams propagating through nonlinear optical materials (Bigelow et al., 2004). The results of this study provide guidance for the selection of the laser beam characteristics to be used to excite nonlinear optical interactions while avoiding the development of laser beam instabilities. In addition, in the course of investigating the nature of nonlinear propagation effects, we uncovered a new mechanism that leads to slow- and fast-light effects in the propagation of light through room-temperature solids.

*Unexpended funds*

There are no uncommitted funds from the prior budget period.

*Publications resulting from this research*:


1. Influence of damping on the vanishing of the electro-optic effect in chiral isotropic media, G.S. Agarwal and R. W. Boyd, Phys. Rev. A, 67 043821, 2003.
2. Observation of Ultra-Slow Light Propagation in a Ruby Crystal at Room Temperature M. S. Bigelow, N. N. Lepeshkin, R. W. Boyd, Phys. Rev. Lett. 90, 113903 (2003).
3. Superluminal and Slow Light Propagation in a Room-Temperature Solid, M. S. Bigelow, N. N. Lepeshkin, and R. W. Boyd, Science, 301, 200, 2003.
4. Thirteen pump-probe resonances of the sodium D1 line by V. Wong, R.W. Boyd, C.R. Stroud Jr., R.S. Bennink, and A.M. Marino, Phys. Rev. A. 68, 012502, 2003.
5. Superluminal and Ultra-Slow Light Propagation in Room-Temperature Solids, R. W. Boyd, M. S. Bigelow, and N. N. Lepeshkin, *Laser Spectroscopy, Proceedings of the XVI International Conference,* pp. 362-364, edited by P. Hannaford, A. Sidorov, H. Bachor, and K. Baldwin, World Scientific Publishing Company, Singapore, 2004.
6. Chirality and Polarization effects in Nonlinear Optics, R. W. Boyd, J. E. Sipe, and P. W. Milonni, Journal of Optics A, 6, S14-S17 (2004).
7. Equivalence of Interaction Hamiltonians in the Electric Dipole Approximation, K. Rzazewski and R.W. Boyd, J. Mod. Optics, 51, 1137 (2004).
8. Influence of Radiative Damping on the Optical-Frequency Susceptibility, P.W. Milonni and R.W. Boyd, Phys. Rev. A, 69, 023814 (2004).
9. Breakup of Ring Beams Carrying Orbital Angular Momentum, M.S. Bigelow, P. Zerom, and R.W. Boyd, Phys. Rev. Lett. 92, 03902, 2004.
10. Measurement of the Intensity-Dependent Refractive Index Using Complete Spatio-Temporal Pulse Characterization, G. Piredda, C. Dorrer, E. M. Kosik, I. A.

Walmsley, and R.W. Boyd, International J. of Nonlinear Optical Physics and Materials, 14, 9 (2005).

11. Transfer of amplitude and phase modulation to a different wavelength using coherently prepared sodium vapor, R. S. Bennink, A. M. Marino, V. Wong, R. W. Boyd, and C. R. Stroud, Jr., Phys. Rev. A **72**, 023827 (2005).
12. T. O. Clatterbuck, P. M. Paul, C. Lynga , B. Sheehy, L. F. DiMauro, P. Agostini, K. C. Kulander and I. A. Walmsley, "Yield and temporal characterization of high-order harmonics from intense midinfrared excitation of a cesium vapor", Phys. Rev. A, 69, 033807 (2004)
13. E. M. Kosik, A. S. Wyatt and L. Corner, E. Cormier, and I. A. Walmsley, 'Spectral phase interferometry for complete reconstruction of attosecond pulses', *Laser Physics* **15**, 909 (2005).
14. E. Cormier, I. A. Walmsley, E. M. Kosik, L. Corner, G. Roberts and L. F. DiMauro, "Self-referencing spatially encoded spectral interferometry for the characterization of attosecond electromagnetic pulses", Phys. Rev. Lett. 94, 033905, (2005)
15. E. Kosik, A. Radunsky, I. A. Walmsley and C. Dorrer, , "Interferometric technique for measuring broadband ultrashort pulses at the sampling limit," Opt. Lett., 30, 326 (2005)
16. E. Kosik, A. Wyatt, L. Corner, E. Cormier and I. A. Walmsley, "Characterization of Attosecond XUV Pulses", Jnl. Mod. Opt., 2005 (invited paper)
17. Adam S. Wyatt, Ian A. Walmsley, Gero Stibenz and Gunter Steinmeyer, 'Spatially encoded arrangement for spectral phase interferometry for direct electric field reconstruction of sub-10 fs pulses', accepted for Optics Letters.

*Conference presentations*

I. A. Walmsley presented an invited talk 'Attosecond Metrology', OSA Annual Meeting, Tucson, AZ, October 2005

I. A. Walmsley presented an invited talk 'Ultrafast Quantum Optics', CLEO, Baltimore, MD, May 2005

I. A. Walmsley presented an invited talk 'Quantum Control', OSA Annual Meeting, Rochester, NY, October 2004

I. A. Walmsley presented an invited talk 'Ultrafast Metrology: Out in the Field', LPHYS'04, Trieste, July, 2004 (Plenary Talk)

I. A. Walmsley presented an invited talk 'Attosecond Metrology', ITAMP workshop, Harvard, November 2003

I. A. Walmsley presented an invited talk ‘Attosecond Metrology’, CLEO, Baltimore, MD, June 2003

V. Wong presented a talk entitled “Saturation-induced extra resonances in pump-probe spectroscopy,” by V. Wong, R. W. Boyd, C. R. Stroud, Jr., and R. S. Bennink at the Topical Meeting on Nonlinear Optics, Co-sponsored by the Optical Society of America and IEEE LEOS, Maui, Hawaii August 2002.

V. Wong presented a talk entitled “Saturation-Induced Extra Resonance in Pump-Probe Spectroscopy,” by V. Wong, R. W. Boyd, C. R. Stroud, Jr., and R. S. Bennink at the OSA Annual Meeting and APS/DLS Laser Science Conference, September 29 – October 3, 2002.

E. Kosik presented a poster entitled “XUV-SPIDER: Toward attosecond pulse measurement” at the Gordon Research Conference on Multi-Photon Processes in July 2002.

*Personnel Visits*

R. W. Boyd visited BNL on June 11, 2002 to present a seminar and to confer with L. DiMauro and other BNL scientists on this project.

R. S. Bennink visited BNL during September 2002 to work with L. DiMauro and other BNL scientists on this project.

E. M. Kosik visited BNL for 6 weeks in August and September 2002 to work with L. DiMauro and P. Colosimo on the BNL HHG source.

L. DiMauro visited Oxford in November 2002 to present a seminar and confer with I. Walmsley and other Oxford scientists on this project.